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(54) HIGHLY HEAT-CONDUCTIVE SHEET AND ITS PREPARATION

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a highly heat-conductive sheet with extremely high heat conductivity as it has good adhesiveness with an electronic instrument and with excellent heat dissipation characteristics.

SOLUTION: The sheet has a gel layer of a heat-conductive filler-contg. silicone resin on one face of a base body sheet consisting of the heat-conductive filler-contg. silicone resin and the max. roughness of the surface of the base body sheet on the opposite side of the gel layer is at most 30 μm and the whole heat conductivity is at least 2.0 W/m.K. As the silicone resin for the matrix of the base body sheet, heat-vulcanizable silicone resins using a peroxide, a room temp.-vulcanizable silicone resins to be vulcanized by condensation reaction, liq. silicone resins to be vulcanizable by addition reaction, etc., are used.

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CLAIMS

[Claim(s)]
[Claim 1] The high temperature conductivity sheet characterized by having the gel layer of thermally conductive filler content silicone resin on one side of the base sheet which consists of thermally conductive filler content silicone resin, for the maximum granularity of the base sheet front face of the opposite side of this gel layer being 30 micrometers or less, and moreover the whole thermal conductivity being 2.0 or more W/m-K.
[Claim 2] The high temperature conductivity sheet according to claim 1 characterized by coming to be reinforced with reinforcing materials.
[Claim 3] The manufacture approach of the high temperature conductivity sheet characterized by coming to contain the following (a) - (d) process.
(a) Silicone resin and the slurry containing a thermally conductive filler The base sheet process [which carries out press cure while the (Process b) above-mentioned unvulcanized base sheet which applies and dries and fabricates an unvulcanized base sheet on a base film had been made to exist on a base film] (c) Obtained is removed from a base film. The process which carries out heating vulcanization and forms a gel layer after applying the silicone gel containing re-pressed (Process d) thermal-conductivity filler and liquefied addition reaction mold silicone resin at least to one side of the base sheet re-pressed [above-mentioned] [claim 4] (b) The manufacture approach of the high temperature conductivity sheet according to claim 3 which the unvulcanized base sheet in a process is the layered product of the unvulcanized base sheet of two or more sheets, and is characterized by moreover stationing reinforcing materials among at least one base sheet of the layered product.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to a high temperature conductivity sheet useful although the heat generated from semiconductor devices in information management systems, such as a computer and a word processor, such as IC, LSI, CPU, and MPU, is emitted efficiently, and its manufacture approach.
[0002]
[Description of the Prior Art] In recent years, as for information management systems, such as a computer and a word processor, the thing of the thin size of portable use is liked. In connection with it, a semiconductor device's increment of densification and the heat which is miniaturized and is generated from there is enhanced, and it has been an important technical problem to remove it efficiently. Conventionally, this heat dissipation is performed by inserting a thermally conductive sheet between electronic equipment and a radiator article, and becoming efficient is also known if the adhesion of a thermally conductive sheet and electronic equipment is raised in this case.
[0003] In order to raise this adhesion, there are some proposals from before. For example, the thermally conductive sheet with which thermal conductivity comes for 10-80, and thickness to prepare [1x10-3 - 5x10-3 cal/cm-sec and ** (0.4 - 2.1 W/m-K), and workability] the silicone gel layer which is 0.05-1.0mm to base material both sides thermal conductivity 1x10-4 cal/cm-sec and more than ** (0.04 or more W/m-K) is indicated by JP,9-17923,A. However, since it is necessary to make the gel layer itself flexible in order to secure adhesion with electronic equipment, by the approach high-filled up with a thermally conductive filler like the advanced technology, adhesion cannot fully be raised, but if thickness of the gel layer which is not high is thickened out of thermal conductivity, the thermal resistance of a thermally conductive sheet will become high.
[0004]
[Problem(s) to be Solved by the Invention] It is offering the high temperature conductivity sheet which this invention's is made in view of the above, and the purpose's has high temperature conductivity, and was moreover excellent in adhesion with electronic equipment, and its manufacture approach.
[0005]
[Means for Solving the Problem] That is, this invention makes the following a summary.
(Claim 1) The high temperature conductivity sheet characterized by having the gel layer of thermally conductive filler content silicone resin on one side of the base sheet which consists of thermally conductive filler content silicone resin, for the maximum granularity of the base sheet front face of the opposite side of this gel layer being 30 micrometers or less, and moreover the whole thermal conductivity being 2.0 or more W/m-K.
(Claim 2) The high temperature conductivity sheet according to claim 1 characterized by coming to be reinforced with reinforcing materials.

(Claim 3) The manufacture approach of the high temperature conductivity sheet characterized by coming to contain the following (a) - (d) process.
(a) Silicone resin and the slurry containing a thermally conductive filler The base sheet process [which carries out press cure while the (Process b) above-mentioned unvulcanized base sheet which applies and dries and fabricates an unvulcanized base sheet on a base film had been made to exist on a base film] (c) Obtained is removed from a base film. The silicone gel containing re-pressed (Process d) thermal-conductivity filler and liquefied addition reaction mold silicone resin Heating vulcanization is carried out after applying at least to one side of the base sheet re-pressed [above-mentioned]. The unvulcanized base sheet in the process (claim 4) (b) process which forms a gel layer The manufacture approach of the high temperature conductivity sheet according to claim 3 which is the layered product of the unvulcanized base sheet of two or more sheets, and is characterized by moreover stationing reinforcing materials among at least one base sheet of the layered product.
[0006]
[Embodiment of the Invention] Hereafter, this invention is further explained to a detail.
[0007] First, explanation of the base sheet which constitutes the high temperature conductivity sheet of this invention uses hot cure mold silicone resin using the peroxide as silicone resin which is the matrix of a base sheet, the room-temperature-vulcanization mold silicone resin vulcanized by the condensation reaction, the liquefied silicone resin vulcanized by the addition reaction.
[0008] Moreover, as a thermally conductive filler which constitutes a base sheet, a kind of metal powder, such as oxide ceramics, such as non-oxide ceramics powder, such as boron nitride, alumimium nitride, silicon carbide, and silicon nitride, and an alumina, silver, copper, and aluminum, or two sorts or more are used. The maximum particle diameter of a thermally conductive filler has desirable 60 micrometers or less. If the maximum particle diameter exceeds 60 micrometers, the maximum surface roughness of a base sheet will surpass 30 micrometers, the adhesion on the front face of a sheet of the side which does not prepare a gel layer will fall, and it will become difficult to perform more efficient heat dissipation.
[0009] Although the amount of the thermally conductive filler used changes with the classes, in order to attain 2.0 or more W/m-K of thermal conductivity of the whole sheet, as for a base sheet, it is desirable that 50 volume % is carrying out thermally conductive filler content at least. The upper limit is 85 volume % extent in consideration of the flexibility of a sheet.
[0010] In this invention, the hardness of base sheet itself is completely arbitrary, and suitable hardness is chosen according to the purpose of using the high temperature conductivity sheet of this invention. For example, about 80 to 100 thing is used by Shore hardness. Adjustment of the hardness of a base sheet can be performed by controlling the class and amount of silicone resin and a thermally conductive filler, hardening extent, etc.
[0011] Although it comes to form the gel layer of a postscript [sheet / of this invention / high temperature conductivity / field / of the above-mentioned base sheet / one], a gel layer is not made to form in the opposite side. Instead, the maximum surface roughness of the opposite side is set to 30 micrometers or less, and the adhesion when including in electronic equipment is raised.
[0012] Next, the gel layer formed in a base sheet front face is explained. the "gel layer" as used in the field of this invention -- a sheet -- toluene -- being immersed -- 5 When it shakes between parts, the part which began to melt is said. since the thickness of a gel layer is dried at the temperature of 80 degrees C after it performs the above-mentioned toluene processing -- weight reduction -- measuring -- a sheet surface product and gel bed density constant:1.52g/cm3 from -- it considers as the computed height value.
[0013] Although what illustrated the silicone resin used for forming a gel layer and a thermally conductive filler by explanation of the above-mentioned base sheet is used, suitable silicone resin is liquefied addition reaction mold silicone resin, and a suitable thermally conductive filler is boron nitride.
[0014] As for the rate of the silicone resin in a gel layer, and a thermally conductive filler, it is desirable that

the thermally conductive filler contains 20 - 45 volume % extent.
[0015] As for especially the thickness of a gel layer, it is desirable that it is about 0.01-0.03mm less than 0.05mm. Even if it becomes rate-limiting in 0.05mm or more radiating heat the thickness of a gel layer by the gel layer and the base sheet itself has high temperature conductivity, more efficient heat dissipation cannot be performed. In addition, as thermal conductivity of the gel layer itself, it is desirable that they are 0.5 or more W/m-K. Moreover, as for the thickness of high temperature conductivity sheet itself of this invention, it is desirable that it is 0.1-1mm.
[0016] The high temperature conductivity sheet of this invention can be made to contain reinforcing materials in the range which is not deviated from the 30 micrometers or less of the maximum surface roughness, and the conditions of 2.0 or more W/m-K of thermal conductivity.
[0017] As reinforcing materials used by this invention, mesh-like insulating materials, metallic foils, etc., such as a glass fiber cross, can be raised. Although the amount used is range which does not remove the conditions of the above-mentioned maximum surface roughness and thermal conductivity, specifically, it is about 15% or less of content among the sheet of a final product.
[0018] Although there is especially no constraint also about a reinforcing materials's existence location, since the effect affect the maximum surface roughness and thermal conductivity by considering as near the center section of the base sheet becomes the smallest, by this invention, it can be called a suitable location.
[0019] Next, the manufacture approach of the high temperature conductivity sheet of this invention is explained. The manufacture approach of this invention suits manufacture of the high temperature conductivity sheet of above-mentioned this invention.
[0020] First, an unvulcanized base sheet is fabricated at the (a) process. Therefore, silicone resin and the slurry of the slurry viscosity 20,000 containing a thermally conductive filler - 60,000cp extent are prepared first. Toluene, a xylene, etc. are used as an organic solvent in that case.
[0021] Subsequently, the above-mentioned slurry is applied and dried on a base film at request thickness. As for a base film, moving continuously is desirable at the point of manufacturing productivity and a uniform sheet, and, as for spreading of a slurry, being based on a doctor blade method is desirable. Although limitation is not received especially in the base film used, also after passing through desiccation and a vulcanization process, what has good detachability with a base sheet is good, for example, the product made of a fluororesin, the product made from polyethylene terephthalate, etc. are suitable.
[0022] Desiccation after slurry spreading is performed at the temperature of about 80 degrees C from a room temperature under an atmospheric-air ambient atmosphere. Since vulcanization is promoted rather than 80 degrees C as it is an elevated temperature, and volatilization of an organic solvent also becomes rapid, pore arises on a base sheet, and thermal conductivity is reduced.
[0023] (b) As for the press cure temperature in a process, it is desirable that it is 40-200 degrees C. At less than 40 degrees C, when a base sheet does not fully vulcanize but surpasses 200 degrees C conversely, there is a possibility that some base sheets may deteriorate.
[0024] press cure is performed under an atmospheric-air ambient atmosphere -- having -- moreover, a press -- for example, a smooth metal -- a wooden floor -- a base sheet -- inserting -- the usual monotonous press machine -- using -- 50 - 150 kgf/cm2 It carries out by the pressure.
[0025] When the high temperature conductivity sheet of this invention is what contains reinforcing materials further, it is desirable to mix it in the phase of this (b) process. As the approach, it is desirable to make it intervene between the unvulcanized base sheets of two or more sheets, and mesh-like insulating materials, metallic foils, etc., such as a glass fiber cross, are suitable as reinforcing materials in that case. Although things, such as powder and a whisker, are satisfactory for the reinforcing materials used by this invention, they are also mixable beforehand in the slurry which in such a case may sprinkle between unvulcanized base sheets and forms a base sheet or a gel layer. Furthermore, compound of these approaches is also possible.
[0026] The (c) process of this invention is a process pressed again, after removing the vulcanized base sheet

from a base film. In the case of this re-press, heat-treatment with a temperature of about 40-150 degrees C may follow if needed.

[0027] (c) a process -- the press to kick -- the bottom of the atmospheric-air ambient atmosphere of a room temperature, and 100 - 500 kgf/cm2 It carries out by the pressure and let the maximum surface roughness of a base sheet be a smooth thing 30 micrometers or less. Only by press processing, when the maximum surface roughness for which it asks cannot be made to reach, auxiliary means, such as a roll press, may be added.

[0028] A gel layer is made to form at least in one side of the base sheet with which the maximum surface roughness was adjusted at the (d) process of this invention. The slurry used here is the mixture containing liquefied addition reaction mold silicone and a thermally conductive filler, and the thing of slurry viscosity 100,000 - 200,000cp extent is used. The viscosity control of liquefied addition reaction mold silicone mainly performs the adjustment. The loadings of a thermally conductive filler can also adjust slurry viscosity. In this case, it is desirable to make that content rate in a gel layer into 20 to 45 volume %, and to perform fine tuning by addition of the above-mentioned organic solvent. If it becomes difficult for the content of the thermally conductive filler in a gel layer to make the thermal conductivity of the whole sheet 2.0 or more W/m-K under by 20 volume % and 45 volume % is surpassed, the hardness of a gel layer will increase and adhesion with electronic equipment will be spoiled.

[0029] Screen-stencil, a roll coater, etc. can perform coating of the slurry for making a gel layer form. Moreover, as for heating vulcanization, it is desirable to carry out using common hot air drying equipment, a far-infrared dryer, a microwave dryer, etc. in the temperature of 100-200 degrees C and 5 - 120 minutes.

[0030]

[Example] Hereafter, an example and the example of a comparison are given and this invention is explained still more concretely.

[0031] 2 volume % was blended with the fill which shows boron nitride powder with a maximum particle diameter of 32 micrometers in Table 1 at example 1 millable type silicone rubber (Toshiba Silicone trade name "TSE221") for toluene, and the slurry of viscosity 10,000cp was prepared. After carrying out coating of this slurry to 0.3mm in thickness on the film made from polyethylene terephthalate using a doctor blade, it put on the hot air drying equipment held at the temperature of 70 degrees C gently for 10 minutes, and the unvulcanized base sheet was fabricated. [(a) Process].

[0032] Two obtained unvulcanized base sheets are inserted with the plate made from stainless steel in piles, and they are the temperature of 150 degrees C, and pressure 100 kgf/cm2. Under the condition, press cure was performed for 45 minutes and the base sheet was manufactured. [(b) Process].

[0033] Subsequently, the base sheet was removed from the film made from polyethylene terephthalate, every one of them was inserted with the plate made from stainless steel this time, and the press was performed for 2 minutes under the room temperature and the condition of pressure 300 kgf/cm2. [(c) Process].

[0034] It is a non-contact type surface roughness meter (after measuring by the trade name "VF-L50" by KEYENCE CORP., according to the following, the gel layer was made to form in that one side, and it considered as the high temperature conductivity sheet of this invention.) about the surface roughness of this re-pressed base sheet. [(d) Process].

[0035] Mix addition reaction mold silicone resin (trade name by Toray Industries Dow Corning "SE1886") 70 volume %, and nitriding hoe prime powder (trade name "DENKA boron nitride" GP grade mean particle diameter of 2 micrometers) 30 volume %, and a slurry is prepared. [by DENKI KAGAKU KOGYO K.K.] After adjusting the viscosity to 120,000cp, it printed in thickness of 0.04mm only at one side of a base sheet using the screen printer furnished with the screen of 75 micrometers of open laps, and vulcanized for 30 minutes in hot air drying equipment with a temperature of 100 degrees C.

[0036] Except having considered as the class and the fill which shows the thermally conductive filler with which each of an example 2 - 5 base sheet, and a gel layer is filled up in Table 1, the high temperature conductivity sheet was produced according to the example 1. However, in the example 2, since the maximum

surface roughness of the base sheet with which it was re-pressed after the (c) process was 40 micrometers, it pressed with the roll press making machine again, and set the maximum surface roughness to 20 micrometers. Moreover, in the example 3, the roll coater performed coating of the slurry of the gel stratification.

[0037] The unvulcanized base sheet with a thickness of 0.1mm was fabricated with the calendering roll using the slurry containing the thermally conductive filler shown in example 6 table 1 [(a) Process]. Aluminium foil with a thickness of 0.04mm is made to intervene between two sheets of this unvulcanized base sheet, and they are the temperature of 150 degrees C, and pressure 80 kgf/cm2. Press cure for 45 minutes under conditions was performed, and the base sheet was manufactured [(b) Process]. The obtained base sheet is removed from the film made from polyethylene terephthalate, it is inserted with the plate made from stainless steel, and it is room temperature and pressure 300 kgf/cm2. The press was performed for 2 minutes under the condition [(c) Process]. Subsequently, the gel layer was formed in one side of this base sheet according to the example 1. However, the roll coater performed printing of a slurry [(d) Process].

[0038] Except having considered as the class and the fill which shows the thermally conductive filler with which each of the example 1 of a comparison - 5 base sheet, and a gel layer is filled up in Table 1, the sheet was produced according to the example 1. However, the base sheet was fabricated in the example 2 of a comparison with the calendering roll.

[0039] About the thermally conductive sheet obtained above, the surface roughness of (1) sheet thickness, (2) gel bed depth, (3) thermal conductivity, and the base sheet of the opposite side in which (4) gel layers were made to form was measured according to the following. Those results are shown in Table 2.

[0040] (1) Sheet thickness : it measured by the micrometer.

(2) gel bed-depth: -- a sheet -- toluene -- being immersed -- 5 a part -- between -- a shaking -- after -- after drying at the temperature of 80 degrees C, weight reduction was measured, height was computed from sheet surface product and gel bed density constant:1.52 g/cm3, and it was made into the gel bed depth.

(3) Thermal conductivity : after inserting the sheet between the TO-3 mold heater case and the copper plate and compressing 10% of sheet thickness, it applied to the copper heater case power 5W, and held for 4 minutes, the temperature gradient of a copper heater case and a copper plate was measured, and thermal conductivity was computed by thermal conductivity (W/m-K) ={(Power W) x thickness (m)}/{(temperature-gradient K) x measurement area (m2)}.

(4) The surface maximum granularity of a base sheet : non-contact type surface roughness meter (it measured by the trade name "VF-L50" by KEYENCE CORP.)

[0041]

[Table 1]

		基体シートの熱伝導性フィラー			備考	ゲル層の熱伝導性フィラー		ゲル層印刷厚み mm
		種類	充填量（体積%）	最大粒子径（μm）		種類	充填量（体積%）	
実施例	1	BN	60	32		BN	30	0.04
	2	Al_2O_3	65	50	再ロールプレス	BN	25	0.03
	3	Si_3N_4	55	43		BN	35	0.04
	4	AlN	52	45		AlN	40	0.02
	5	SiO_2	65	29		AlN	30	0.02
	6	BN	60	32	Al箔補強	BN	30	0.04
比較例	1	BN	40	45		BN	40	0.04
	2	AlN	60	120		BN	45	0.03
	3	BN	58	32		BN	50	0.10
	4	BN	58	32		BN	60	0.02
	5	Si_3N_4	65	25		BN	15	0.02

[0042]
[Table 2]

		シート厚み（mm）	ゲル層厚み（mm）	熱伝導率（W/m・K）	表面最大粗さ（μm）
実施例	1	0.28	0.03	3.4	26
	2	0.36	0.03	3.0	20
	3	0.41	0.04	3.7	21
	4	0.37	0.01	3.6	21
	5	0.43	0.02	3.1	25
	6	0.63	0.03	3.1	23
比較例	1	0.50	0.03	1.6	18
	2	0.41	0.03	1.5	51
	3	0.51	0.09	1.6	26
	4	0.32	0.02	1.9	26
	5	0.48	0.02	1.3	28

[0043]
[Effect of the Invention] Since the high temperature conductivity sheet of this invention has good adhesion with electronic equipment, its thermal conductivity is very high, and it is excellent in a heat dissipation property.

[Translation done.]